

Gapless spin liquid and valence-bond solid in the J_1 - J_2 Heisenberg model on the square lattice: Insights from singlet and triplet excitations

Francesco Ferrari¹ and Federico Becca²

¹*Institute for Theoretical Physics, Goethe University Frankfurt, Max-von-Laue-Straße 1, D-60438 Frankfurt am Main, Germany*

²*Dipartimento di Fisica, Università di Trieste, Strada Costiera 11, I-34151 Trieste, Italy*

The spin-1/2 J_1 - J_2 Heisenberg model on the square lattice represents one of the simplest examples in which the effects of magnetic interactions may suppress magnetic order, eventually leading to a pure quantum phase with no local order parameters. This model has been extensively studied in the last three decades, with conflicting results. Here, by using Gutzwiller-projected wave functions and recently developed methods to assess the low-energy spectrum, we show the existence of a level crossing between the lowest-energy triplet and singlet excitations for $J_2/J_1 \approx 0.54$. This fact supports the existence of a phase transition between a gapless spin liquid (which is stable for $0.48 \lesssim J_2/J_1 \lesssim 0.54$) and a valence-bond solid (for $0.54 \lesssim J_2/J_1 \lesssim 0.6$), even though no clear sign of dimer order is visible in the correlations functions. These results, which confirm recent density-matrix renormalization calculations on cylindrical clusters [L. Wang and A. W. Sandvik, *Phys. Rev. Lett.* **121**, 107202 (2018)], reconcile the contradicting results obtained within different approaches over the years.

I. INTRODUCTION

Since the beginning of the field of highly frustrated magnetism, the spin-1/2 Heisenberg model, defined on the square lattice with both nearest- (J_1) and next-nearest-neighbor (J_2) superexchange couplings [1], has played a pivotal role in the search for and characterization of unconventional states of matter, in which correlation effects cannot be neglected [2]. In the J_1 - J_2 model, the combination of quantum and geometrical frustrations may lead to a ground state that has no local order parameters (such as a finite magnetization) and hosts elementary excitations with fractional quantum numbers. While the limiting case with $J_2 = 0$ (and, equivalently, $J_1 = 0$) can be assessed by Monte Carlo techniques and shows a conventional ground state with finite magnetization [3–5], in the presence of a finite frustrating J_2 term, there are no approaches that can give unbiased results on large lattices. Many different numerical techniques have been used to understand the nature of the ground state in the vicinity of $J_2/J_1 = 0.5$, where the highest level of frustration is expected (in the classical limit, where spins are treated as vectors of fixed length, this point presents a large degeneracy in the lowest-energy manifold).

In early studies, some evidence of a valence-bond solid was obtained, from analytical approximations [6] and numerical calculations on small clusters [7,8]. Later, this scenario was confirmed by series expansion techniques [9]. On the other hand, subsequent Monte Carlo calculations with Gutzwiller-projected fermionic wave functions hinted at the possibility that the ground state of the system may be a pure quantum spin liquid, with no dimer order [10]. More recently, several works addressed the question of the ultimate nature of the ground-state wave function in the highly frustrated region $0.5 \lesssim J_2/J_1 \lesssim 0.6$, with contradicting outcomes, supporting the existence of a valence-bond solid (with either columnar

or plaquette order) or a spin liquid (either gapped or gapless) [11–25]. In this respect, recent developments in the numerical optimization of the wave function with many parameters, such as tensor- or neural-network states, open promising routes to further investigate this delicate issue [26–29].

Remarkably, in a recent paper on the subject, Wang and Sandvik [30] suggested the possibility that the nonmagnetic region of the model may consist of two phases: a gapless spin liquid, which develops continuously after the Néel state, and a valence-bond solid, which is stabilized for larger values of the frustrating ratio [30]. In particular, by applying the density-matrix renormalization group technique to $2L \times L$ cylinders, they showed the existence of a level crossing between the lowest-energy triplet and singlet excitations for $J_2/J_1 \approx 0.52$. Since the Néel order is expected to disappear for $J_2/J_1 \gtrsim 0.46$, this feature has been associated with the transition between a gapless spin liquid and a valence-bond solid; likewise, in the one-dimensional J_1 - J_2 model, a similar level crossing at $J_2/J_1 \approx 0.24$ marks the transition between the gapless (critical) phase and the dimerized one [31,32].

Here, we assess the issue of the level crossing between the lowest-energy triplet and singlet within variational wave functions constructed by using Abrikosov fermions, subject to the Gutzwiller projection that enables us to work in the correct Hilbert space of the spin model [33]. Besides the standard calculations for the ground-state wave function, we apply the variational method proposed by Li and Yang [34] and recently developed by us [24,35] to construct both low-energy triplet and singlet states. The main motivation is to give an independent validation of the claim of Wang and Sandvik, considering, instead of $2L \times L$ cylindrical geometries (with open boundary conditions along the x direction), $L \times L$ square clusters with periodic boundary conditions, thus retaining all the symmetries of the infinite lattice. This fact allows us to

identify the momenta of the excitations, which are $q = (\pi, \pi)$ for the triplet and $q = (\pi, 0)$ [or $q = (0, \pi)$] for the singlet. In addition, the singlet with $q = (\pi, \pi)$ remains at much higher energies, thus suggesting that the valence-bond order is presumably columnar and not staggered or plaquette (these two cases require such a singlet in the ground-state manifold). In our previous work on the dynamical structure factor [24], evidence of gapless triplet excitations up to $J_2/J_1 \approx 0.55$ was reported, supporting the existence of a spin liquid with Dirac-like spinon excitations (and \mathcal{Z}_2 gauge excitations) [17]. Then, the main outcome of the present work is to confirm the presence of a singlet-triplet level crossing, which appears for $J_2/J_1 \approx 0.54$, in excellent agreement with what was obtained by Wang and Sandvik [30]. However, within the clusters that are presently available (i.e., L up to 20), a precise size-scaling extrapolation of the gaps is extremely difficult, not excluding a vanishing value in the thermodynamic limit in the entire nonmagnetic phase. In this regard, dimer-dimer correlations of the ground state are compatible with a power-law decay both before and after the level crossing, implying a very large correlation length and a tiny dimer-order parameter. Therefore, it turns out that, as for the one-dimensional J_1 - J_2 model, the phase transition is much more easily detected by looking at the level crossing, instead of looking at ground-state properties or at the triplet gap alone. Still, we mention that our calculations (as well as the ones performed by Wang and Sandvik) cannot exclude that the level crossing in two dimensions, instead of signaling the onset of a valence-bond solid, could mark an unconventional transition between two paramagnetic phases, both supporting gapless singlet and triplet excitations.

II. MODEL AND METHODS

The Hamiltonian of the J_1 - J_2 Heisenberg model on the square lattice is defined by

$$\mathcal{H} = J_1 \sum_{\langle R, R' \rangle} \mathbf{S}_R \cdot \mathbf{S}_{R'} + J_2 \sum_{\langle\langle R, R' \rangle\rangle} \mathbf{S}_R \cdot \mathbf{S}_{R'}, \quad (1)$$

where $\langle \dots \rangle$ and $\langle\langle \dots \rangle\rangle$ indicate pairs of nearest- and next-nearest-neighbor sites, respectively. The phase diagram of the system features two magnetic phases: for $J_2/J_1 \lesssim 0.48$ the ground state displays Néel order, while for $J_2/J_1 \gtrsim 0.6$ a different magnetic order is established, in which the spins align ferromagnetically in one direction and antiferromagnetically in the other direction.

In this work, we focus on the nonmagnetic region $0.48 \lesssim J_2/J_1 \lesssim 0.6$, and we approximate the ground state and the excited states of the system by means of variational wave functions based on Gutzwiller-projected fermions. To formulate our variational guess for the ground state of the spin system, we introduce an auxiliary BCS Hamiltonian of Abrikosov fermions,

$$\mathcal{H}_0 = \sum_{R, R', \sigma} t_{R, R'} c_{R, \sigma}^\dagger c_{R', \sigma} + \sum_{R, R'} \Delta_{R, R'} c_{R, \uparrow}^\dagger c_{R', \downarrow}^\dagger + \text{H.c.}, \quad (2)$$

and we apply the Gutzwiller projector $\mathcal{P}_G = \prod_R n_R(2 - n_R)$ to its ground state $|\Phi_0\rangle$. The Gutzwiller projection suppresses all the fermionic configurations with empty or doubly occupied sites, thus returning a suitable wave function for spins,

namely, $|\Psi_0\rangle = \mathcal{P}_G |\Phi_0\rangle$. In the nonmagnetic region of the J_1 - J_2 model the best variational *Ansatz* for the ground state is obtained by a BCS Hamiltonian with an s -wave hopping at first neighbors, a $d_{x^2-y^2}$ pairing at first and fourth neighbors, and a d_{xy} pairing at fifth neighbors [10,17]. The optimal values of the hopping and pairing parameters are obtained by minimizing the variational energy with the stochastic re-configuration technique [36]. We note that $|\Psi_0\rangle$ is a singlet state which possesses all the symmetries of the lattice [33], and we emphasize that any possible attempt to break translational symmetry within the auxiliary BCS Hamiltonian (e.g., including a dimerized hopping or pairing) does not lead to any energy gain with respect to the uniform *Ansatz* (in contrast to the one-dimensional case [37]).

Following the idea of Li and Yang [34], the fermionic formalism can also be employed to design variational *Ansätze* for the excited states of the spin model. The variational scheme is based on the definition of a set of projected particle-hole excitations:

$$|q, R\rangle_\pm = \mathcal{P}_G \sum_{R'} e^{iqR'} (c_{R+R', \uparrow}^\dagger c_{R', \uparrow} \pm c_{R+R', \downarrow}^\dagger c_{R', \downarrow}) |\Phi_0\rangle. \quad (3)$$

Here, $|q, R\rangle_+$ and $|q, R\rangle_-$ are, respectively, singlet and triplet states with momentum q , which are labeled by the Bravais lattice vector R . Taking suitable linear combinations of these states, we can accurately approximate the low-energy singlet and triplet excitations of the spin model. In particular, the coefficients of the linear combinations are determined by the Rayleigh-Ritz variational principle, i.e., by diagonalizing the Hamiltonian of the J_1 - J_2 model within the subset $\{|q, R\rangle_+\}_R$ for the singlet sector and $\{|q, R\rangle_-\}_R$ for the $S^z = 0$ triplet sector [37]. The level crossing is analyzed by looking at the lowest-energy triplet and singlet wave functions constructed in this way.

III. RESULTS

In order to benchmark our variational calculations, we consider the one-dimensional J_1 - J_2 model. In this case, the transition between the gapless phase (stable for small J_2/J_1) and the gapped dimerized one (stable for large J_2/J_1) has been located by looking at the singlet-triplet level crossing [31,32], with a very high level of accuracy of the transition point [38,39]. In the one-dimensional case, both triplet and singlet excitations have $q = \pi$ (with respect to the ground state). The variational wave function used in this case is the same as the one that was used in our previous calculations [37], which is constructed from an auxiliary BCS Hamiltonian that contains a first-neighbor hopping plus on-site and second-neighbor pairings. In Fig. 1, we report the gaps for a small cluster with $L = 30$ (where exact diagonalizations are available) and for a large one with $L = 94$. The comparison with exact results proves the accuracy of our estimations of the variational gaps, showing that the phase transition appears in the correct place also when increasing the lattice size (the aim of this calculation is not to compete with previous estimations of the critical value). The size scaling of the gaps confirms gapless excitations for small frustrating ratios; by contrast, inside the dimerized phase, the triplet is gapped, while the singlet is collapsing to the ground state exponentially (the thermodynamic value is

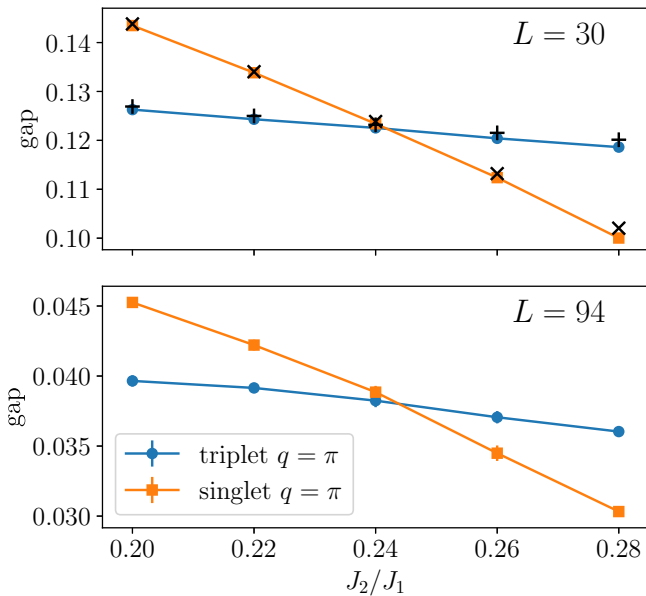


FIG. 1. Triplet and singlet gaps for $L = 30$ (top) and $L = 94$ (bottom) for different values of the frustrating ratio J_2/J_1 in the one-dimensional J_1 - J_2 model. The exact results for $L = 30$ are also shown for comparison (black crosses).

consistent with zero within a few error bars). These results are shown in Fig. 2. In the vicinity of the transition, the triplet gap is exponentially small, and therefore, it is extremely hard to detect a finite value from an unbiased size scaling. In this respect, the transition is much better located by looking at the level crossing.

Being confident that our variational method is able to reproduce the correct features of the lowest-energy triplet and singlet excitations, we move to the most interesting two-dimensional model. First of all, we report in Fig. 3 our results for the $2L \times L$ geometry used in Ref. [30] (the only difference is that, here, we consider periodic boundary conditions in both directions, suitable for a translational-invariant wave function). Here, we find a level crossing between the triplet

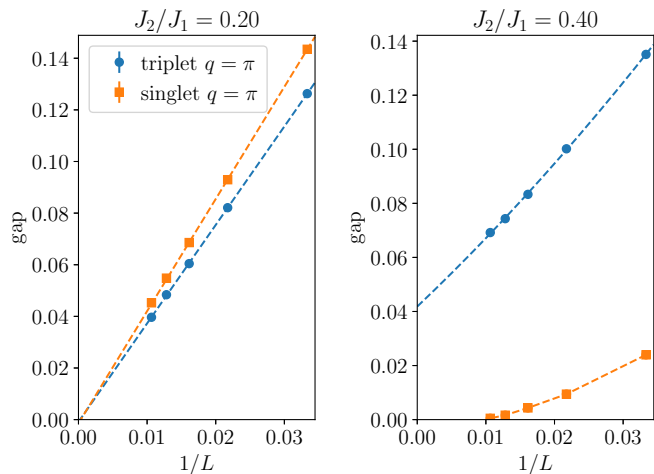


FIG. 2. Size scaling of the triplet and singlet gaps of the one-dimensional J_1 - J_2 model for two values of J_2/J_1 .

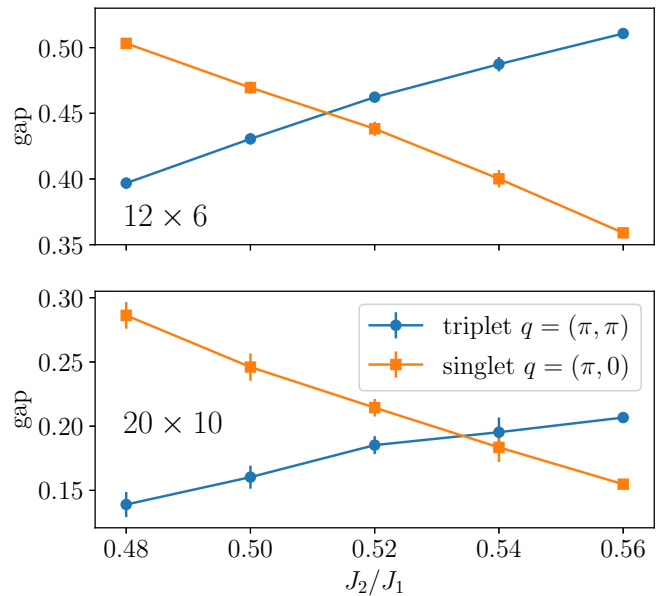


FIG. 3. Triplet [with $q = (\pi, \pi)$] and singlet [with $q = (\pi, 0)$] gaps for $2L \times L$ clusters with $L = 6$ (top) and $L = 10$ (bottom) for different values of the frustrating ratio J_2/J_1 .

with $q = (\pi, \pi)$ and the singlet with $q = (\pi, 0)$, similar to what was obtained within the density-matrix renormalization group. We would like to mention that, within this geometry, the crossing point moves from $J_2/J_1 \approx 0.51$ for $L = 6$ to $J_2/J_1 \approx 0.535$ for $L = 10$, in qualitative agreement with Ref. [30]. Similar results can be obtained within $L \times L$ clusters (see Fig. 4). The advantage of this kind of geometry, besides having all the point-group symmetries of the square lattice, is that the crossing point does not move substantially when changing the value of L . The size scaling of the

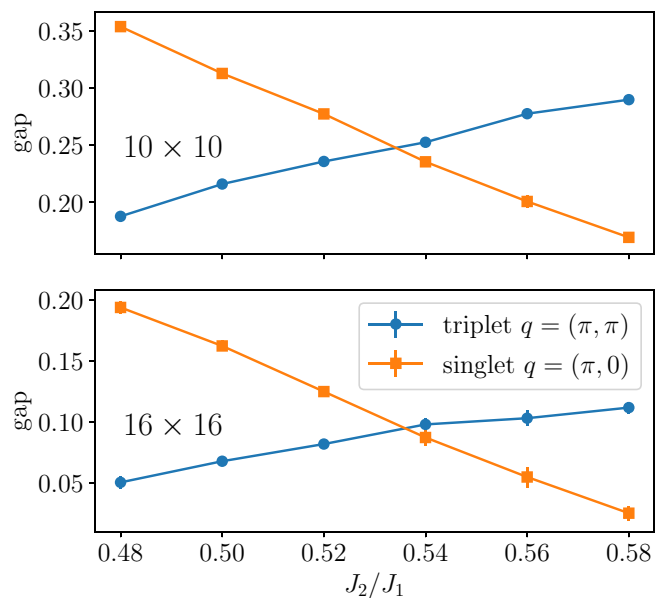


FIG. 4. The same as in Fig. 3 for $L \times L$ clusters with $L = 10$ (top) and $L = 16$ (bottom).

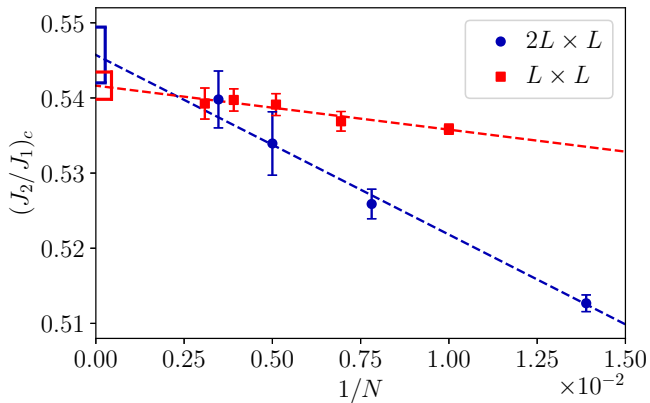


FIG. 5. Size scaling of the singlet-triplet crossing point $(J_2/J_1)_c$ for $2L \times L$ and $L \times L$ geometries. Data are extrapolated as a function of $1/N$, where N is the total number of sites (analogous to what is done in Ref. [30]).

crossing point is reported in Fig. 5 for both $2L \times L$ and $L \times L$ clusters. The best fit is obtained with the latter choice, giving $(J_2/J_1)_c = 0.542(2)$, while the former one gives $(J_2/J_1)_c = 0.546(4)$. These estimates are slightly different from the one obtained in Ref. [30], which found $(J_2/J_1)_c \approx 0.52$. This discrepancy may be ascribed either to the approximate nature of our variational approach or to issues on the convergence of the density-matrix renormalization group procedure. We would like to emphasize that a direct evaluation of the thermodynamic gap is extremely difficult in the two-dimensional case since the asymptotic regime is not reached within the available clusters (with L up to 18). By contrast, the examination of the level crossing gives very stable results, with tiny size effects, demonstrating the clear advantages of this diagnostic to assess quantum phase transitions.

Considering that the magnetic Néel order vanishes at $J_2/J_1 \approx 0.48$ [24], the present results are compatible with the existence of a spin-liquid region in its vicinity, i.e., for $0.48 \lesssim J_2/J_1 \lesssim 0.54$. Beyond that, it is reasonable to expect a different phase, presumably with columnar dimer order. This conclusion is corroborated by the fact that the lowest-energy singlet excitations have $q = (\pi, 0)$ and $q = (0, \pi)$. The possibility of staggered dimers or a plaquette valence-

bond order is not probable since the ground-state manifold would also include a singlet at $q = (\pi, \pi)$, which instead lies much higher in energy.

With the aim of assessing the properties of the variational state $|\Psi_0\rangle$, we computed the isotropic spin-spin $\langle \mathbf{S}_0 \cdot \mathbf{S}_R \rangle$ and dimer-dimer $\langle D_0^\mu D_R^\mu \rangle$ correlations [$D^\mu(R) = \mathbf{S}_R \cdot \mathbf{S}_{R+\hat{\mu}}$, with $\mu = x$ or y]. The results are reported in Fig. 6 for $J_2/J_1 = 0.5$ and 0.58 , which lie in the two different regions, sufficiently far away from the crossing point. Remarkably, the behavior is similar in both cases (at least for the largest cluster used in this work, i.e., 48×48). The spin-spin correlations have a power-law decay, with exponents that are consistent with 2, namely, $2.0(1)$ for $J_2/J_1 = 0.5$ and $2.1(1)$ for $J_2/J_1 = 0.58$. We remark that density-matrix renormalization group calculations [30] obtained different exponents of this putative power-law behavior, suggesting that further investigations in this direction are needed to fully characterize the nature of the ground-state wave function. Also the dimer-dimer correlations do not show an appreciable difference between these two values of J_2 , displaying a power-law decay in both cases (in this case, evaluating the exponent is much harder, given the rapid decay of the signal). At first glance, these results may suggest an extremely large correlation length that persists in the whole region where dimer order is expected from the level-crossing analysis. Alternatively, it is possible that the *Ansatz* used for this region can be improved, including an explicit dimer order (as we discussed above, we could not find any simple way to include a symmetry breaking that provides a variational energy gain). This hypothetical wave function may display much clearer evidence for dimerization, even in the correlation functions. In this respect, we want to mention that in one spatial dimension the level crossing can be (accurately) detected also using a simple wave function with only nearest-neighbor hopping, which has power-law dimer-dimer correlations (not shown). However, as discussed above, it is possible to have a substantial energy gain by including pairing terms in the fermionic Hamiltonian and to obtain a wave function with finite dimer-dimer correlations at long distances without affecting the level crossing between the lowest-energy triplet and singlet. In this regard, we definitively believe that our results on the level crossing in the two-dimensional case are peculiar features of the model and do not depend crucially on the details of the wave function, provided sufficiently

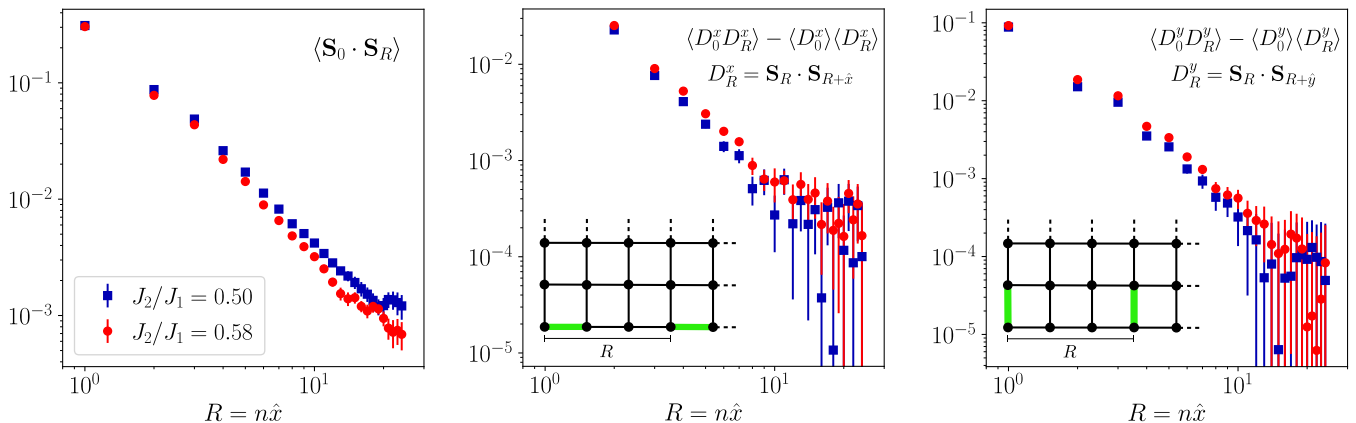


FIG. 6. Spin-spin and dimer-dimer correlations on the 48×48 cluster for $J_2/J_1 = 0.5$ and $J_2/J_1 = 0.58$.

accurate *Ansätze* are considered. On the other hand, we note that detecting the valence-bond ordered phase by just looking at the ground-state properties is a complicated task since the dimer order is limited to a very narrow region of the phase diagram.

IV. CONCLUSIONS

By using a variational approach based on Gutzwiller-projected fermionic wave functions, we showed the existence of a level crossing between the lowest-energy triplet, with $q = (\pi, \pi)$, and singlet, with $q = (\pi, 0)$ and $(0, \pi)$, excitations within the paramagnetic region of the J_1 - J_2 Heisenberg model on the square lattice. Our results are in excellent agreement with the recent density-matrix renormalization group calculation by Wang and Sandvik [30], with a tiny difference in locating the Néel-to-spin-liquid transition ($J_2/J_1 = 0.48$ vs 0.46) and the singlet-triplet level crossing ($J_2/J_1 = 0.54$ vs 0.52). Most likely, the level crossing indicates the existence of a critical point separating the gapless spin liquid and a

valence-bond solid with columnar order [a staggered dimer order, as well as a plaquette valence-bond order, can be ruled out by observing that the lowest singlet with $q = (\pi, \pi)$ remains high in energy]. Remarkably, within the variational *Ansatz* used in this work (on clusters up to 48×48) spin-spin correlations display a power-law behavior also beyond the level crossing; in addition, no visible order parameter is seen from dimer-dimer correlations. These facts may be explained by an extremely large correlation length. Even if we could not exclude the possibility that the level crossing marks a more unconventional phase transition, we emphasize that looking to the low-energy spectrum represents a very powerful and valuable tool to establish the ultimate phase diagram of frustrated spin models.

ACKNOWLEDGMENTS

We thank L. Wang and A. Sandvik for useful discussions. F.F. acknowledges support from the Alexander von Humboldt Foundation.

-
- [1] P. Chandra and B. Doucot, *Phys. Rev. B* **38**, 9335(R) (1988).
 - [2] L. Balents, *Nature (London)* **464**, 199 (2010).
 - [3] J. D. Reger and A. P. Young, *Phys. Rev. B* **37**, 5978 (1988).
 - [4] A. W. Sandvik, *Phys. Rev. B* **56**, 11678 (1997).
 - [5] M. Calandra Buonaura and S. Sorella, *Phys. Rev. B* **57**, 11446 (1998).
 - [6] N. Read and S. Sachdev, *Phys. Rev. Lett.* **62**, 1694 (1989); *Phys. Rev. B* **42**, 4568 (1990).
 - [7] F. Figueirido, A. Karlhede, S. Kivelson, S. Sondhi, M. Rocek, and D. S. Rokhsar, *Phys. Rev. B* **41**, 4619 (1990).
 - [8] H. J. Schulz, T. A. L. Ziman, and D. Poilblanc, *J. Phys. I France* **6**, 675 (1996).
 - [9] R. R. P. Singh, Z. Weihong, C. J. Hamer, and J. Oitmaa, *Phys. Rev. B* **60**, 7278 (1999).
 - [10] L. Capriotti, F. Becca, A. Parola, and S. Sorella, *Phys. Rev. Lett.* **87**, 097201 (2001).
 - [11] O. P. Sushkov, J. Oitmaa, and Z. Weihong, *Phys. Rev. B* **63**, 104420 (2001).
 - [12] M. Mambrini, A. Lauchli, D. Poilblanc, and F. Mila, *Phys. Rev. B* **74**, 144422 (2006).
 - [13] J. Richter and J. Schulenburg, *Eur. Phys. J. B* **73**, 117 (2010).
 - [14] H.-C. Jiang, H. Yao, and L. Balents, *Phys. Rev. B* **86**, 024424 (2012).
 - [15] F. Mezzacapo, *Phys. Rev. B* **86**, 045115 (2012).
 - [16] L. Wang, D. Poilblanc, Z.-C. Gu, X.-G. Wen, and F. Verstraete, *Phys. Rev. Lett.* **111**, 037202 (2013).
 - [17] W.-J. Hu, F. Becca, A. Parola, and S. Sorella, *Phys. Rev. B* **88**, 060402(R) (2013).
 - [18] S.-S. Gong, W. Zhu, D. N. Sheng, O. I. Motrunich, and M. P. A. Fisher, *Phys. Rev. Lett.* **113**, 027201 (2014).
 - [19] R. L. Doretto, *Phys. Rev. B* **89**, 104415 (2014).
 - [20] S. Morita, R. Kaneko, and M. Imada, *J. Phys. Soc. Jpn.* **84**, 024720 (2015).
 - [21] D. Poilblanc and M. Mambrini, *Phys. Rev. B* **96**, 014414 (2017).
 - [22] R. Haghshenas and D. N. Sheng, *Phys. Rev. B* **97**, 174408 (2018).
 - [23] W.-Y. Liu, S. Dong, C. Wang, Y. Han, H. An, G.-C. Guo, and L. He, *Phys. Rev. B* **98**, 241109(R) (2018).
 - [24] F. Ferrari and F. Becca, *Phys. Rev. B* **98**, 100405(R) (2018).
 - [25] M. Hering, J. Sonnenschein, Y. Iqbal, and J. Reuther, *Phys. Rev. B* **99**, 100405(R) (2019).
 - [26] H.-J. Liao, J.-G. Liu, L. Wang, and T. Xiang, *Phys. Rev. X* **9**, 031041 (2019).
 - [27] K. Choo, T. Neupert, and G. Carleo, *Phys. Rev. B* **100**, 125124 (2019).
 - [28] F. Ferrari, F. Becca, and J. Carrasquilla, *Phys. Rev. B* **100**, 125131 (2019).
 - [29] A. Szabó and C. Castelnovo, [arXiv:2002.04613](https://arxiv.org/abs/2002.04613).
 - [30] L. Wang and A. W. Sandvik, *Phys. Rev. Lett.* **121**, 107202 (2018).
 - [31] K. Okamoto and K. Nomura, *Phys. Lett. A* **169**, 433 (1992).
 - [32] G. Castilla, S. Chakravarty, and V. J. Emery, *Phys. Rev. Lett.* **75**, 1823 (1995).
 - [33] X.-G. Wen, *Phys. Rev. B* **65**, 165113 (2002).
 - [34] T. Li and F. Yang, *Phys. Rev. B* **81**, 214509 (2010).
 - [35] F. Ferrari and F. Becca, *Phys. Rev. X* **9**, 031026 (2019).
 - [36] S. Sorella, *Phys. Rev. B* **71**, 241103(R) (2005).
 - [37] F. Ferrari, A. Parola, S. Sorella, and F. Becca, *Phys. Rev. B* **97**, 235103 (2018).
 - [38] S. Eggert, *Phys. Rev. B* **54**, R9612 (1996).
 - [39] A. W. Sandvik, in *Lectures on the Physics of Strongly Correlated Systems XIV: Fourteenth Training Course in the Physics of Strongly Correlated Systems*, AIP Conf. Proc. No. 1297 (AIP, New York, 2010), p. 135.